Supporting Information for "Relativistic effects in Photon-Induced Near Field Electron Microscopy"

Sang Tae Park and Ahmed H. Zewail*
Physical Biology Center for Ultrafast Science and Technology,
Arthur Amos Noyes Laboratory of Chemical Physics,
California Institute of Technology, Pasadena, California 91125
(Dated: May 9, 2012)

I. DIRAC EQUATION

The Dirac equation for an electron in an electromagnetic wave is given by

$$i\hbar\frac{\partial\Psi}{\partial t}=\left\{c\boldsymbol{\alpha}\cdot\left(\vec{p}-q\vec{A}\right)+q\phi+\boldsymbol{\beta}mc^2\right\}\Psi \tag{S.1}$$

where $q=-e$ is the electron charge, $\vec{A}$ is the vector potential, and ϕ is the scalar potential. $\boldsymbol{\alpha}$ and $\boldsymbol{\beta}$ are unit constant matrices chosen to satisfy the relativistic energy-momentum relation. For a one dimensional equation, along the z-direction, $\boldsymbol{\alpha}$, $\boldsymbol{\beta}$, and Ψ are given by two components. as

$$\boldsymbol{\alpha} = \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix} \tag{S.2}$$

$$\boldsymbol{\beta} = \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix} \tag{S.3}$$

$$\Psi = \begin{pmatrix} \psi_1 \\ \psi_2 \end{pmatrix} \tag{S.4}$$

and eqn. (S.1) becomes

$$i\hbar\frac{\partial}{\partial t}\begin{pmatrix} \psi_1 \\ \psi_2 \end{pmatrix}=\begin{pmatrix} q\phi+mc^2 & c\left(\hat{p}_z-qA_z\right) \\ c\left(\hat{p}_z-qA_z\right) & q\phi-mc^2 \end{pmatrix}\begin{pmatrix} \psi_1 \\ \psi_2 \end{pmatrix} \tag{S.5}$$

II. FOURIER TRANSFORM OF GAUSSIAN FUNCTION

Fourier transformation for differentiation shows the following property:

$$\mathcal{F}^{-1}\left\{i^n k^n \hat{F}\left(k\right)\right\}=\left(\frac{d}{dz}\right)^n\mathcal{F}\left\{\hat{F}\left(k\right)\right\} \tag{S.6}$$

The Gaussian profiles in momentum and position spaces are defined as,

$$\hat{G}\left(k;\sigma_k\right) = \frac{1}{\sqrt{2\pi}\sigma_k}\exp\left[-\frac{k^2}{2\sigma_k^2}\right] \tag{S.7}$$

$$G\left(z;\sigma_z\right) = \frac{1}{\sqrt{2\pi}\sigma_z}\exp\left[-\frac{z^2}{2\sigma_z^2}\right] \tag{S.8}$$

then with $\sigma_z=\frac{1}{2\sigma_k}$, they satisfy

$$\mathcal{F}^{-1}\left\{\sqrt{\hat{G}\left(k-k_0;\sigma_k\right)}\right\} = \sqrt{G\left(z;\sigma_z\right)}e^{ik_0z} \tag{S.9}$$

$$\mathcal{F}^{-1}\left\{i\left(k-k_0\right)\sqrt{\hat{G}\left(k-k_0;\sigma_k\right)}\right\} = \frac{d\sqrt{G\left(z;\sigma_z\right)}}{dz}e^{ik_0z} \tag{S.10}$$

*zewail@caltech.edu

III. RELATIVISTIC PINEM SOLUTION

As in the non-relativistic case, the vector potential of the scattered wave by linearly polarized incident wave is given by

$$A_z = \frac{1}{2}\left(\tilde{A_z} + \tilde{A_z}^{\dagger}\right) = \frac{1}{2}\left(\frac{\tilde{E_z}}{i\omega_p} - \frac{\tilde{E_z}^{\dagger}}{i\omega_p}\right) = \frac{1}{\omega_p} Im\left[\tilde{E_z}\right] \tag{S.11}$$

The temporal dependence of the scattered electric field can be separated as $\tilde{E}\left(z,t\right) = \tilde{E}\left(z,0\right)\exp\left[-i\omega_p t\right]$ for a continuous wave. For an incident optical pulse of duration much longer than the electron transit time, the scattered wave in the vicinity of the nanostructure can be approximated as

$$\tilde{E}\left(z,t\right) \approx \tilde{E}\left(z,0\right)\exp\left[-i\omega_p t\right]\exp\left[-\frac{\left(t-\tau\right)^2}{4\sigma_p^2}\right] \tag{S.12}$$

Then, we obtain

$$\frac{g^{+}\left(z',t\right)}{g^{+}\left(z',t_0\right)} = \exp\left[+i\frac{qv_0}{\hbar\omega_p}\int_{t_0}^{t} dt' Im\left\{\tilde{E}_z\left(z'+v_0t',0\right)\exp\left[-i\omega_p t'\right]\exp\left[-\frac{\left(t'-\tau\right)^2}{4\sigma_p^2}\right]\right\}\right] \tag{S.13}$$

By substituting $z'' = z' + v_0 t'$ in the integration and using the fact that $\tilde{E}\left(z'',0\right)$ is only significant around $z'' = 0$, we derive

$$\begin{aligned}\frac{g^{+}\left(z',t\right)}{g^{+}\left(z',t_0\right)} &= \exp\left[+i\frac{q}{\hbar\omega_p}\int_{z'+v_0t_0}^{z'+v_0t} dz'' Im\left\{\tilde{E}_z\left(z'',0\right)\exp\left[-i\omega_p\frac{z''-z'}{v_0}\right]\exp\left[-\frac{\left(z''-z'-v_0\tau\right)^2}{4v_0^2\sigma_p^2}\right]\right\}\right] \\ &\approx \exp\left[+i\frac{q}{\hbar\omega_p} Im\left\{\exp\left[i\frac{\omega_p}{v_0}z'\right]\int_{z'+v_0t_0}^{z'+v_0t} dz''\tilde{E}_z\left(z'',0\right)\exp\left[-i\frac{\omega_p}{v_0}z''\right]\exp\left[-\frac{\left(-z'-v_0\tau\right)^2}{4v_0^2\sigma_p^2}\right]\right\}\right]\end{aligned}$$

By substituting $t = +\infty$ and $t_0 = -\infty$, the final state can be expressed as

$$\frac{g^{+}\left(z',+\infty\right)}{g^{+}\left(z',-\infty\right)} = \exp\left[+i\frac{q}{\hbar\omega_p} Im\left\{\exp\left[i\frac{\omega_p}{v_0}z'\right]\int_{-\infty}^{+\infty} dz''\tilde{E}_z\left(z'',0\right)\exp\left[-i\frac{\omega_p}{v_0}z''\right]\right\}\exp\left[-\frac{\left(z'+v_0\tau\right)^2}{4v_0^2\sigma_p^2}\right]\right] \tag{S.14}$$

Using the definition of $\tilde{F} = \tilde{F}\left(\frac{\omega_p}{v_0}\right) = \int_{-\infty}^{+\infty} dz''\tilde{E}_z\left(z'',0\right)\exp\left[-i\left(\frac{\omega_p}{v_0}\right)z''\right]$, eqn. (S.14) becomes

$$\frac{g^{+}\left(z',+\infty\right)}{g^{+}\left(z',-\infty\right)} = \exp\left[+i\frac{q}{\hbar\omega_p} Im\left\{\exp\left[i\frac{\omega_p}{v_0}z'\right]\tilde{F}\right\}\exp\left[-\frac{\left(z'+v_0\tau\right)^2}{4v_0^2\sigma_p^2}\right]\right] \tag{S.15}$$

The electron part in eqn. (S.15) is identical to eqn. (A12) in the previous publication [1], and no relativistic correction is required when the relativistic velocity is used for both formulations, (and corresponding k_c and ω_c for the classical formulation). Similarly, we Taylor-expand the exponential functions in eqn. (S.15), re-substitute Im function by subtraction of its complex conjugate, and rearrange it using the definition of a Bessel function (Jacobi-Anger relation) to obtain

$$\frac{g^{+}\left(z',+\infty\right)}{g^{+}\left(z',-\infty\right)} = \sum_{n=-\infty}^{+\infty}\exp\left[in\frac{\omega_p}{v_0}z'\right]\left(\frac{-\tilde{F}}{\left|\tilde{F}\right|}\right)^{n} J_n\left(\frac{-q}{\hbar\omega_p}\left|\tilde{F}\right|\exp\left[-\frac{\left(z'+v_0\tau\right)^2}{4v_0^2\sigma_p^2}\right]\right) \tag{S.16}$$

IV. FINAL DIRAC WAVEFUNCTION

We define the spinor vector as

$$\hat{\mathbf{u}}^{+}_{(k)} = \begin{pmatrix} u_1^{+}\left(k\right) \\ u_2^{+}\left(k\right) \end{pmatrix} \tag{S.17}$$

$$\hat{\mathbf{u}}^{-}_{(k)} = \begin{pmatrix} u_1^{-}\left(k\right) \\ u_2^{-}\left(k\right) \end{pmatrix} \tag{S.18}$$

and define $\xi_n(z') = \left(-\frac{\tilde{F}}{|\tilde{F}|}\right)^n J_n\left(\frac{-q}{\hbar\omega_p}\left|\tilde{F}\right| \exp\left[-\frac{(z'+v_0\tau)^2}{4v_0^2\sigma_p^2}\right]\right)$ and $g_n^+(z') = g^+(z', -\infty)\,\xi_n(z')$, such that

$$g^+(z', +\infty) = \sum_{n=-\infty}^{+\infty} \exp\left[in\frac{\omega_p}{v_0}z'\right] g_n^+(z') \tag{S.19}$$

The final state wavefunction is retrieved by substituting $f^+(z,t) = g^+(z - v_0 t, t)$ and $f^-(z,t) \approx \frac{1}{-2i\omega_0}\frac{c}{\gamma_0}\left(-\frac{\partial f^+(z,t)}{\partial z}\right)\exp\left[-i\left(\omega_0^+ - \omega_0^-\right)t\right]$ in eqn. (17) of the *main text* at $t \to +\infty$, where $\hbar\omega_0 = \gamma_0 mc^2$, such that

$$\Psi(z,t) = \left\{g^+(z', +\infty)\,\hat{\mathbf{u}}^+_{(k_0)} - \frac{i\hbar}{2\gamma_0^2 mc}\frac{\partial g^+(z', +\infty)}{\partial z}\hat{\mathbf{u}}^-_{(k_0)}\right\}\exp\left[i\left(k_0 z - \omega_0^+ t\right)\right] \tag{S.20}$$

which becomes

$$\Psi(z,t) = \sum_{n=-\infty}^{+\infty}\left\{\exp\left[in\frac{\omega_p}{v_0}z'\right] g_n^+(z')\,\hat{\mathbf{u}}^+_{(k_0)} - \frac{i\hbar}{2\gamma_0^2 mc}\frac{\partial\left(\exp\left[in\frac{\omega_p}{v_0}z'\right] g_n^+(z')\right)}{\partial z}\hat{\mathbf{u}}^-_{(k_0)}\right\}\exp\left[i\left(k_0 z - \omega_0^+ t\right)\right] \tag{S.21}$$

where the differential term becomes

$$\frac{\partial\left(\exp\left[in\frac{\omega_p}{v_0}z'\right] g_n^+(z')\right)}{\partial z} = in\frac{\omega_p}{v_0}\exp\left[in\frac{\omega_p}{v_0}z'\right] g_n^+(z') + \exp\left[in\frac{\omega_p}{v_0}z'\right]\frac{\partial}{\partial z}g_n^+(z') \tag{S.22}$$

Eqn. (S.21) becomes

$$\begin{aligned}
\Psi(z,t) &= \sum_{n=-\infty}^{+\infty}\left\{g_n^+(z')\,\hat{\mathbf{u}}^+_{(k_0)} + g_n^+(z')\left(n\frac{\omega_p}{v_0}\right)\frac{\hbar}{2\gamma_0^2 mc}\hat{\mathbf{u}}^-_{(k_0)} - \frac{\partial g_n^+(z')}{\partial z}\frac{i\hbar}{2\gamma_0^2 mc}\hat{\mathbf{u}}^-_{(k_0)}\right\}\exp\left[in\frac{\omega_p}{v_0}z'\right]\exp\left[i\left(k_0 z - \omega_0^+ t\right)\right]\\
&= \sum_{n=-\infty}^{+\infty}\left\{g_n^+(z')\left(\hat{\mathbf{u}}^+_{(k_0)} + n\frac{\omega_p}{v_0}\frac{\hbar}{2\gamma_0^2 mc}\hat{\mathbf{u}}^-_{(k_0)}\right) - \frac{\partial g_n^+(z')}{\partial z}\frac{i\hbar}{2\gamma_0^2 mc}\hat{\mathbf{u}}^-_{(k_0)}\right\}\exp\left[in\frac{\omega_p}{v_0}z'\right]\exp\left[i\left(k_0 z - \omega_0^+ t\right)\right]\\
&= \sum_{n=-\infty}^{+\infty}\left\{g_n^+(z')\,\hat{\mathbf{u}}^+_{(k_n)} - \frac{\partial g_n^+(z')}{\partial z}\frac{i\hbar}{2\gamma_0^2 mc}\hat{\mathbf{u}}^-_{(k_0)}\right\}\exp\left[in\frac{\omega_p}{v_0}(z - v_0 t)\right]\exp\left[i\left(k_0 z - \omega_0^+ t\right)\right]\\
&= \sum_{n=-\infty}^{+\infty}\left\{g_n^+(z')\,\hat{\mathbf{u}}^+_{(k_n)} - \frac{\partial g_n^+(z')}{\partial z}\frac{i\hbar}{2\gamma_0^2 mc}\hat{\mathbf{u}}^-_{(k_0)}\right\}\exp\left[i\left(k_n z - \omega_n^+ t\right)\right]\\
&\approx \sum_{n=-\infty}^{+\infty}\left\{g_n^+(z')\,\hat{\mathbf{u}}^+_{(k_n)} - \frac{\partial g_n^+(z')}{\partial z}\frac{i\hbar}{2\gamma_0^2 mc}\left(\hat{\mathbf{u}}^-_{(k_0)} - n\frac{\omega_p}{v_0}\frac{\hbar}{2\gamma_0^2 mc}\hat{\mathbf{u}}^+_{(k_0)}\right)\right\}\exp\left[i\left(k_n z - \omega_n^+ t\right)\right]\\
&= \sum_{n=-\infty}^{+\infty}\left\{g_n^+(z')\,\hat{\mathbf{u}}^+_{(k_n)} - \frac{\partial g_n^+(z')}{\partial z}\frac{i\hbar}{2\gamma_0^2 mc}\hat{\mathbf{u}}^-_{(k_n)}\right\}\exp\left[i\left(k_n z - \omega_n^+ t\right)\right]\\
&= \sum_{n=-\infty}^{+\infty}\left\{g_n^+(z')\,\hat{\mathbf{u}}^+_{(k_n)} - i\frac{\partial g_n^+(z')}{\partial z}\left.\frac{\partial\hat{\mathbf{u}}^+_{(k)}}{\partial k}\right|_{k=k_n}\right\}\exp\left[i\left(k_n z - \omega_n^+ t\right)\right]
\end{aligned} \tag{S.23}$$

where $k_n = k_0 + n\frac{\omega_p}{v_0}$, and $\omega_n = \omega_0 + n\omega_p$. An approximation of $\hat{\mathbf{u}}^-_{(k_n)} \approx \hat{\mathbf{u}}^-_{(k_0)}$ was used in the small range of k limit.

In the momentum space, the wavefunction becomes

$$\begin{aligned}
\Psi(k) &\approx \sum_{n=-\infty}^{+\infty}\mathcal{F}\left\{g_n^+(z - v_0 t)\exp\left[i\left(k_n z - \omega_n^+ t\right)\right]\left\{\hat{\mathbf{u}}^+_{(k_0)} + (k - k_0)\left.\frac{\partial\hat{\mathbf{u}}^+_{(k)}}{\partial k}\right|_{k=k_n}\right\}; k\right\} && \text{(S.24)}\\
&\approx \sum_{n=-\infty}^{+\infty}\mathcal{F}\left\{g_n^+(z - v_0 t)\exp\left[i\left(k_n z - \omega_n^+ t\right)\right]; k\right\}\begin{pmatrix} u_1^+(k) \\ u_2^+(k)\end{pmatrix} && \text{(S.25)}\\
&= \sum_{n=-\infty}^{+\infty}\mathcal{F}\left\{g_n^+(z'); k - k_n\right\}\begin{pmatrix} u_1^+(k) \\ u_2^+(k)\end{pmatrix} && \text{(S.26)}
\end{aligned}$$

The probability of each wavelet becomes

$$P_n = \int_{k_n-\frac{1}{2}\frac{\omega_p}{v_0}}^{k_n+\frac{1}{2}\frac{\omega_p}{v_0}} dk \left| \sum_{n=-\infty}^{+\infty} \mathcal{F}\left\{g_n^+; k-k_n\right\} \right|^2 \tag{S.27}$$

$$\approx \int_{k_n-\frac{1}{2}\frac{\omega_p}{v_0}}^{k_n+\frac{1}{2}\frac{\omega_p}{v_0}} dk \left|\mathcal{F}\left\{g_n^+; k-k_n\right\}\right|^2 \tag{S.28}$$

$$\approx \int_{-\infty}^{+\infty} dk \left|\mathcal{F}\left\{g_n^+; k-k_n\right\}\right|^2 \tag{S.29}$$

when each wavelet does not overlap with the other, as $\mathcal{F}\left\{g_n^+; k-k_n\right\}\mathcal{F}\left\{g_m^+; k-k_m\right\} \approx 0$. Eqn. (S.29) is equivalent to the classical counterpart:

$$\Psi(k) = \sum_{n=-\infty}^{+\infty} \mathcal{F}\left\{g_n; k-k_n\right\} \tag{S.30}$$

$$P_n \approx \int_{-\infty}^{+\infty} dk \left|\mathcal{F}\left\{g_n; k-k_n\right\}\right|^2 = \int_{-\infty}^{+\infty} dz' \left|g_n(z')\right|^2 \tag{S.31}$$

V. RELATIVISTIC MASS

Relativistic transverse and longitudinal masses are

$$m_T = \gamma m = \frac{p}{v} \tag{S.32}$$

$$m_L = \gamma^3 m = \frac{F}{a} = \frac{\partial p}{\partial v} \tag{S.33}$$

VI. CLASSICAL EQUIVALENCE

The Schrödinger formulation of electromagnetic interaction is relativistically valid when classical momentum and energy are used as

$$\hbar k_c = p_c = m v_0 = \frac{p_0}{\gamma_0} \tag{S.34}$$

$$\hbar\omega_c = T_c = \frac{1}{2} m v_0^2 = \frac{\left(\gamma_0^2-1\right) mc^2}{2\gamma_0^2} = \frac{\gamma_0+1}{2\gamma_0^2} T_0 \tag{S.35}$$

or when the momentum with the relativistic (transverse) mass, $m_T = \gamma_0 m$, and corresponding energy are used as

$$\hbar k_c = p_c = m_T v_0 = \gamma_0 m v_0 = p_0 \tag{S.36}$$

$$\hbar\omega_c = T_c = \frac{1}{2} m_T v_0^2 = \frac{\left(\gamma_0^2-1\right)\gamma_0 mc^2}{2\gamma_0^2} = \frac{\gamma_0+1}{2\gamma_0} T_0 \tag{S.37}$$

In both cases, eqn. (4) in the *main text* is identical to eqn. (24), and eqn. (4) does not require a relativistic correction, as long as the actual velocity, v_0, is used. Correct usage of parameters in various formulations are summarized in Table S.1.

---

[1] S. T. Park, M. Lin, and A. H. Zewail, New J. Phys. **12**, 123028 (2010).

TABLE S.1: Comparison of formulations

	m	v_i	$k_i\ (p_i/\hbar)$	$\omega_i\ (E_i/\hbar)$	Δk	$\Delta\omega$
Experiment[a]	m_e	$v_0 = 0.695c$	$k_0 = 0.967 m_e c/\hbar$	$\omega_0 = 1.391 m_e c^2/\hbar$	$\frac{\omega_p}{v_0}$	ω_p
Dirac formalism	m_e	v_0	k_0	ω_0	$\frac{\omega_p}{v_0}$	ω_p
Schrödinger formalism	m_e	v_0	$k_c = 0.695 m_e c/\hbar$[b]	$\omega_c = 0.242 m_e c^2/\hbar$[c]	$\frac{\omega_p}{v_0}$	ω_p
Effective mass picture	$m_T = 1.391 m_e$[d]	v_0	k_0	$\omega_T = 0.336 m_e c^2/\hbar$[e]	$\frac{\omega_p}{v_0}$	ω_p

[a]For the experiment, $\gamma_0 = 1 + \frac{T_0}{m_e c^2}$ is evaluated, and one obtains $v_0 = c\frac{\sqrt{\gamma_0^2-1}}{\gamma_0}$, $p_0 = \hbar k_0 = m_e c\sqrt{\gamma_0^2 - 1}$, and $E_0 = \hbar\omega_0 = \gamma_0 m_e c^2$.
[b]$k_c = \frac{m v_0}{\hbar}$
[c]$\omega_c = \frac{m_e v_0^2}{2\hbar}$
[d]$m_T = \gamma_0 m_e$
[e]$\omega_T = \frac{\gamma_0 m_e v_0^2}{2\hbar}$